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"Lusaka, Zambia, during SAFARI-2000: A Collection Point for Ozone Pollution"

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We present the first set of ozone soundings in the heart of the southern African burning region, which were taken during 6-11 Sept 2000 during the SAFARI-2000 campaign. Up to now, all ozone sondes have recorded ozone pollution from intense southern African burning at more remote continental or Island sites, mostly at Ascension in the Atlantic and Reunion Island in the Indian Ocean. Surprising results! (1) Local burning practices (roadside clearing, charcoal manufacture) contributed to 95 ppbv ozone surface readings; (2) column tropospheric ozone was > 50 DU, much higher than concurrent measurements over Nairobi and Pretoria SHADOZ stations; (3) the absolutely stable pollution layers that circulate over southern Africa (first detected south of 20S during SAFARI-92) appear to be a more general feature of southern African dynamics; (4) the heaviest pollution layer (800-500 hPa) over Lusaka is due to recirculated trans-boundary ozone. Starting out over western Zambia, Angola, and Namibia, ozone heads east to the Indian Ocean, before wrapping around the southern African gyre, and turning back over Mozambique and Zimbabwe, ending up over Lusaka. This makes Lusaka a collection point for pollution.

Lusaka, Zambia, during SAFARI-2000: A Collection Point for Ozone Pollution

April 26, 2002-For Submittal to GRL

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Abstract. In August and September, throughout south central Africa, seasonal clearing of dry vegetation and other fire-related activities lead to intense smoke haze and ozone formation. The first ozone soundings in the heart of the southern African burning region were taken at Lusaka, Zambia (15.5S, 28E) in early September 2000. Over 90 ppbv ozone was recorded at the surface (1.3 km elevation) and column tropospheric ozone was > 50 DU during a stagnant period. These values are much higher than concurrent measurements over Nairobi (1S, 38E) and Irene (25S, 28E, near Pretoria). The heaviest ozone pollution layer (800-500 hPa) over Lusaka is due to recirculated trans-boundary ozone. Starting out over Zambia, Angola, and Namibia, ozone heads east to the Indian Ocean, before turning back over Mozambique and Zimbabwe, heading toward Lusaka. Thus, Lusaka is a collection point for pollution, consistent with a picture of absolutely stable layers recirculating in a gyre over southern Africa.

1. Introduction: Southern African Ozone and Circulation.

Savanna burning is a well-known source of pollutants that lead to ozone formation. During SAFARI-92 (Southern African Fires Atmospheric Reserach Initiative, September 1992) emission factors over southern African savanna fires were deduced [Andreae, 1997] and ozone budgets were determined from ozone soundings and aircraft measurements [Thompson et al., 1996]. Analysis of transport over southern Africa revealed the persistence of anticyclonic flow and extremely stable pollution layers [Garstang and Tyson, 1997; Tyson et al., 1997]. The term "absolutely stable layers" refers to extreme atmospheric stability [Tyson et al., 1997], including elevated inversions. Vertical mixing is inhibited though intact layers may rise and fall day-to-day. The lifetime of ozone above the boundary layer may exceed several weeks [Thompson et al., 1996]. In that case, ozone may be fairly wellconserved during advection. Indeed, ozone laminae traceable to Africa are well-known over the Pacific [Newell et al., 1999; Oltmans et al., 2001].

During SAFARI-2000, the first ozone soundings made in the heart of south central African burning (at Lusaka, Zambia, 15.5S, 28E) offered an opportunity to determine

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whether stable layers are a more general feature of the region (Figure 1). Other goals of the measurements were: absolute measurement of column ozone for evaluation of satellite ozone measurements; comparison of Lusaka ozone profiles with those at routinely operating African stations: Irene (25S, 28E) and Nairobi, Kenya (1S, 38E).

In this paper we show that stable layers were indeed present at Lusaka and that local pollution contributes to very high surface ozone values (> 90 ppbv). Even higher ozone mixing ratios were in an absolutely stable layer at 2-5 km (800-500 hPa) that was transported toward Lusaka by easterly winds from the Indian Ocean, Zimbabwe and Mozambique. Wrapped around an anticyclonic high-pressure system, the ozone ultimately originated from firerich rural areas of Botswana, South Africa, Namibia, Angola, and western Zambia, making Lusaka a collection point for regional pollution.

2. In-situ and Remotely Sensed Measurements. Total Ozone Column Comparisons.

Ozone and temperature profiles (recorded at 1-s frequency) were determined with an electrochemical concentration cell ozonesonde (ENSCI2Z) in combination with an RS-80/15 Väisäla radiosonde and a HumiCap humidity sensor, as described in Thompson et al. [2000]. The procedures for sonde preparation and data acquisition were developed by NOAA/Climate Diagnostics and Monitoring Laboratories [Johnson et al., 2002]. Launches were made at the Zambian Meteorological Department (ZMD) in Lusaka (15.5S, 28E, 1.3 km elevation) between 6 and 11 September 2000. Two launches were made on 7, 9 and 10 September 2000. Soundings were recorded to an 8-10 hPa balloon burst; the first 7 September 2000 sounding lost sonde signal at ~40 hPa when laboratory power failed. AOT (aerosol optical thickness) and total overhead ozone were measured at 1-2 hour intervals from 1-11 September 2000 with a multichannel sun photometer (Solar Light Co.) The 380-nm AOT data are used in this paper. TOMS (Total Ozone Mapping Spectrometer) total ozone data are taken from satellite overpasses closest to Lusaka (shortly before local noon).

Total and tropospheric ozone column amounts (in Dobson Units; $1 DU = 2.69 \times 10^{16} \text{ molec/cm}(3)$), integrated from the soundings, are shown as a time-series in Figure 2. AOT from the sun photometer is also displayed. Agreement between TOMS total ozone (a in Figure 2) and total ozone integrated from the sondes is excellent. In five of six days, agreement ranges from + 7 DU (sonde total ozone greater than TOMS) to - 3 DU (TOMS greater than sonde total). The sun photometer ozone readings (5-7 per day, not shown) were also in good agreement with sonde and satellite total ozone, bracketing the TOMS and sonde ozone totals on most days. The good comparison between TOMS total ozone at Lusaka and the sonde total is similar to the mean agreement (1-2%) recorded at Irene and Nairobi [Thompson et al., 2002a]. All three sites have > 1 km elevation, which minimizes one source of TOMS error, a diminished detection efficiency for ozone in the lower troposphere [Hudson et al., 1995].

Figure 2 shows the effect of a localized disturbance that occurred in Lusaka early on 9 September. High surface wind speeds and a shift in wind direction from easterly to northwesterly led to a reduction in total and tropospheric ozone. There was a simultaneous decrease in AOT. Between two soundings taken 4 hours apart on 9 September, tropospheric ozone column declined 10 DU, from 49 DU to 39 DU (Figure 2).

3. Absolutely Stable Ozone Layers. Origins of Lusaka Ozone.

Using radiosonde data taken over 5 South African cities and Windhoek, Namibia during September 2000, Freiman et al. [2002] showed that stable layers, similar to those observed during SAFARI-92, persisted on 80% or more of the days in August and September 2000. Stable layers appear in the Lusaka ozonesondes, with all nine profiles showing appreciable pollution (> 35 DU tropospheric ozone) and at least one layer with ozone > 70 ppbv. Five sets of the ozone-humidity profiles are displayed in Figures 3 and 4. There is a fairly consistent location of pollution layers in relative humidity and temperature as well as in For example, prior to the 9 September disturbance, stagnant air with a pronounced inversion at 780 hPa (temperature at 2.2 km in Figure 3) allows an absolutely stable ozone layer (> 120 ppbv) to develop (lower highlighted band, Figure 3). Between 8 and 10 km (temperature, Figure 3) there is a second layer of ozone > 80 ppbv. Although diminished in ozone and drier, the lower stable layer remained intact in the 10 and 11 September profiles, one of which is displayed in Figure 4.

Where does the ozone over Lusaka come from? On examining the meteorological conditions prevailing during the Lusaka sampling, Freiman and Riphagen [2002] show that surface flows were predominantly easterly. This is reflected in a cluster of trajectories shown arriving at Lusaka at 900 hPa on 7 September (orange in Figure 5). The ozone profile for 7 September is illustrated in Figure 4. Aloft, at 700 hPa (blue in Figure 5) trajectories reveal the typical SAFARI-2000 pattern of anticyclonic flow about a subsiding gyre [Jury and Freiman, 2002]. Air parcels at this level, ~2 km (lower highlighted band in Figure 4), passed over fires occurring throughout southern Africa (Figure 5). The arrival at Lusaka is southeasterly from the Indian Ocean, passing over Mozambique and Zimbabwe (Figure 1), with origins 4-5 days earlier over the Atlantic and fire-rich areas of Angola, Namibia, Botswana, and South Africa. The 500 hPa air parcels (green in Figure 5), corresponding to the upper part of the stable layer in the ozone soundings on 6-8 September 2000 (Figures 3 and 4), were over Angola and western Zambia prior to arrival over Lusaka. Although elevated ozone mixing ratios near the surface at Lusaka are partly of local origin (below), ozone aloft is imported from burning regions over surrounding countries.

4. Tropospheric Ozone Profiles over Lusaka, Nairobi, Irene.

The Lusaka soundings show very high tropospheric ozone (54 DU, integrated tropospheric column) during 6-8 September (Figure 2). Figure 3 depicts high ozone mixing ratios throughout the troposphere on 6 September. Near-surface ozone averaged 90 ppbv and within a double temperature inversion layer (3-5 km), ozone was > 120 ppbv. Ozone maxima in the boundary layer and in the inversion layer immediately above coincide with layers of elevated relative humidity (Figure 4). A drier layer from 5.5-10 km is nearly uniform in ozone (~75 ppbv) and a second tropospheric maximum layer (95 ppbv) occurs at 12 km.

Low level trajectories (900 hPa in Figure 5) showed that some of the boundary layer ozone at Lusaka appears to be of local origin. In addition to routine trash fires and motor vehicle pollution, organized burning along roadsides (Figure 6) and seasonal charcoal making were ongoing during the sampling period. For several hours after the ozone sounding of 8 September, a second sonde was operated at the ground at ZMD (1500-1700 local time). A steady 95 ppbv mixing ratio was observed; this is the highest surface ozone value reported to date over southern Africa. The following morning (9 September), surface ozone had dropped to 55 ppbv (Figure 4) as a result of the surface disturbance that caused a 10 DU tropospheric ozone decrease from the 8th (Figure 1). Two days later (11 September), with the return of stagnant conditions, surface ozone during the morning launch was 80 ppbv. If the ozone buildup is ascribed to local sources, the formation rate is-12 ppbv/day, similar to ozone formation rates deduced from aircraft sampling near active fires in SAFARI-92/TRACE-A (Transport and Atmospheric Chemistry near the Equator-Atlantic; Thompson et al., 1996).

Figure 3 shows two other southern African ozone soundings on 6 September 2000. These profiles display less pollution than over Lusaka. Nairobi pollution on 6 September was concentrated in peaks centered at 9 and 13 km (green profile). Because there was relatively little burning over east central Africa during September 2000 (fire counts in Figure 5), the high-ozone layers over Nairobi were probably transported several hundred km from sources. The blue profile (Irene) in Figure 3 is one of the most polluted profiles in the 1998-2000 observing period at this site (Figure 13 in *Thompson et al.*, 2002*b*). Figure 5 suggests that biomass burning in South Africa and Mozambique may contribute to elevated ozone at Irene, an interpretation consistent with local meteorology [Freiman et al., 2002; Freiman and Riphagen, 2002].

5. Summary

The first ozone soundings over Zambia, taken during a typical dry-season burning period in September 2000, showed high ozone concentrations throughout the troposphere. Surface concentrations of 95 ppbv at Lusaka, the highest readings measured to date in southern Africa, were much greater than surface ozone at

Nairobi and Irene (Pretoria) at the same time. Satellite total column ozone from TOMS agreed well with the sondes and with column measurements from a portable ozone uv-photometer. Free tropospheric ozone over Lusaka was concentrated in absolutely stable layers, a phenomenon that characterizes southern African circulation more generally. The two most prominent layers over Lusaka in early September 2000 were at 3-5 km and 8-12 km. The lower of these layers appears to be a mixture of predominantly imported origins. Specifically, ozone from western Zambia and neighboring countries is exported, sometimes via the Indian Ocean, before returning to the continent. Lusaka is thus a collecting point for recirculated trans-boundary pollution within the southern African gyre.

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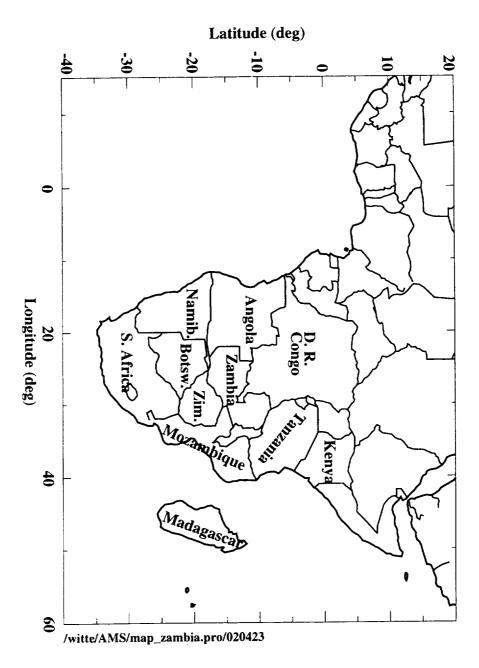
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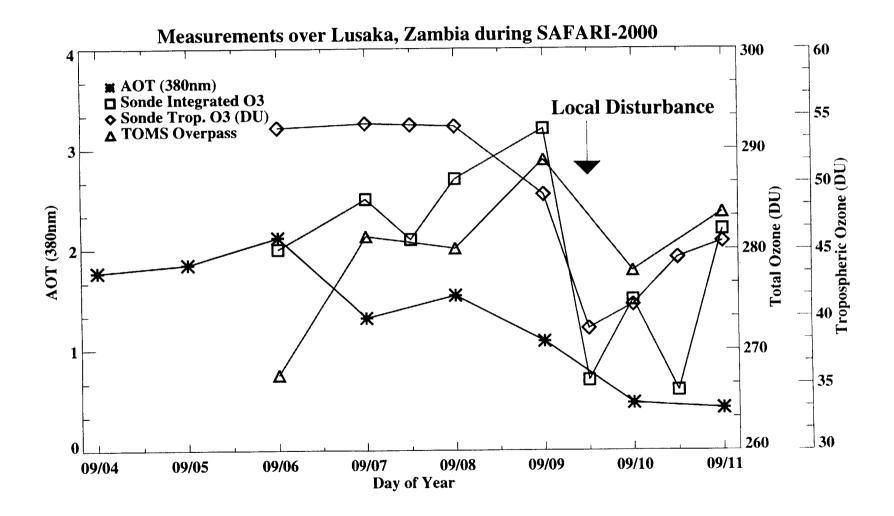
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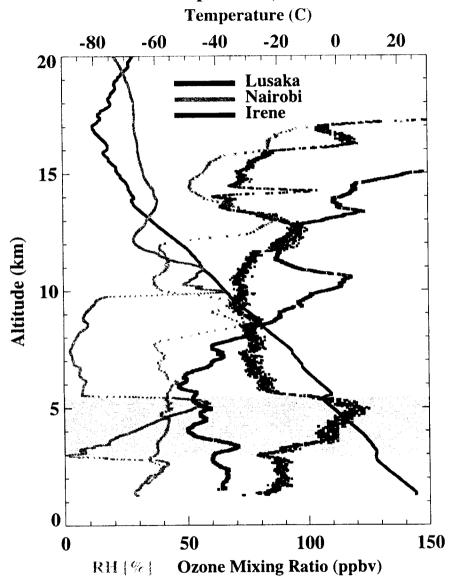
FIGURE CAPTIONS

- Figure 1. Major countries in southern Africa.
- Figure 2. Time-series of aerosol optical thickness (from handheld sun photometer, 380 nm channel); total, tropospheric ozone integrated from the Lusaka soundings, and total ozone from TOMS overpasses during SAFARI-2000 early September 2000 sampling. A stagnant period led to gradual increases in AOT and satellite total ozone from 1-6 September (not shown). Similar variations were observed in readings taken over Mongu, 400 km west of Lusaka (T. Eck, personal communication, 2001), indicating that the Lusaka conditions are representative of the larger region during this period (see also Freiman et al., 2002). A disturbance in surface winds, observed in radiosondes launched by ZMD at Lusaka (not shown) is indicated with an arrow. Between the two soundings on 9 September, tropospheric ozone decreased 10 DŪ and stratospheric ozone declined by > 10 DU. The latter difference may not be statistically significant given the precision of the sonde stratospheric ozone [Thompson et al., 2002a]. Mean stratospheric ozone for the 8 Lusaka soundings that reached 10 hPa was 230+/- 5 DU.
- **Figure 3.** Profiles of ozone, temperature and relative humidity from launches on 6 September 2000 over Lusaka, Zambia. Ozone profiles from 6 September launches at Irene and Nairobi are also illustrated.
- **Figure 4.** Ozone and relative humidity profiles over Lusaka 7-10 September 2000. Shading shows absolutely stable layers as classified by *Freiman et al.* [2002]. On 9 Sept. a disturbance diluted surface ozone and some ozone aloft. By 11 September stagnant conditions returned and ozone and aerosols built up again (Fig. 2).
- Figure 5. AVHRR fire counts (red dots) for September 2000, courtesy of http://shark1.esrin.esa.it/ionia/FIRE. Five-day back trajectory of parcels from Lusaka area, initialized at 500, 700, and 900 hPa. Clusters of back trajectories were run for five days using a kinematic version of the Goddard trajectory model [Schoeberl and Newman, 1995] to capture uncertainties associated with analyzed winds (NCEP 2.5 x 2.5 deg). Time of trajectory initiation is 12Z on 7 Sept 2000. These transport patterns are confirmed by trajectories run with winds [Freiman and Riphagen, 2002] from the higher resolution Eta model [0.5 deg, three-hourly data; Black, 1994]. For transport to Mongu, 400 km west of Lusaka, two-day origins were from Botswana and Namibia, with an anticyclonic change in direction over Zimbabwe [Freiman and Piketh, 2002].
- Figure 6. Photo of organized burning in downtown Lusaka, ~200 m from ZMD ozonesonde launch site. Crews set fires to clear debris on both sides of roadway. Each day the crews burn several hundred meters along the road.

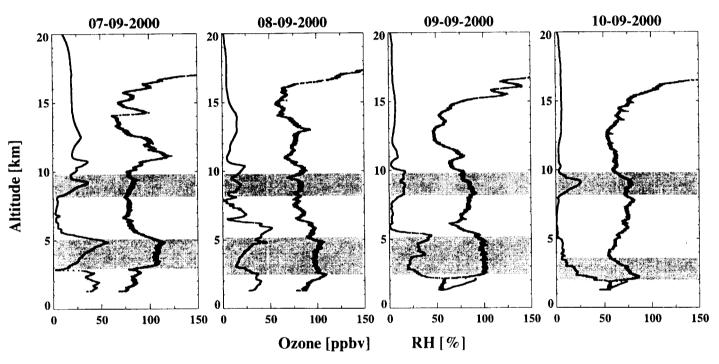




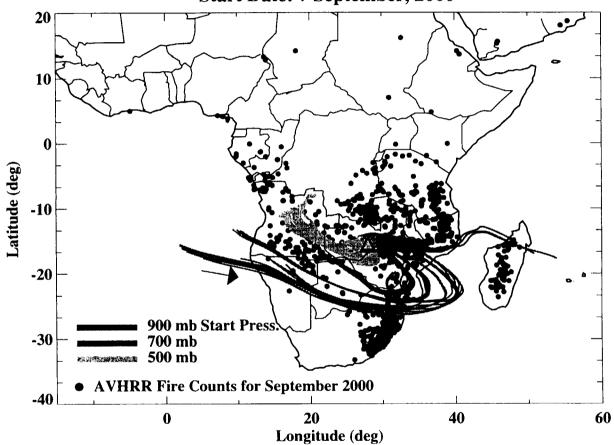
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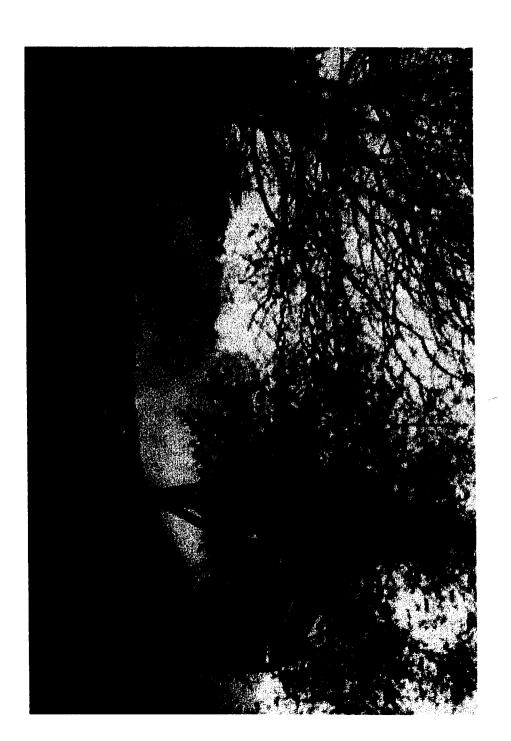


Ozonesonde Profiles at Lusaka, Zambia



5-day Back-trajectories arriving over Lusaka, Zambia Start Date: 7 September, 2000





Flgure 6